Neutron Resonance Spectroscopy: The Application of Neutron Physics to Shock and Material Physics

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Introduction

Neutron resonance spectroscopy (NRS) has developed a technique that uses Doppler-broadened neutron resonances to take fast snapshots of internal temperatures in dynamically loaded samples. The use of neutron resonances to measure temperatures in static samples was first pioneered at the Los Alamos Neutron Science Center (LANSCE) by British experimenters in the mid 1980s. The experimenters were working in conjunction with the Rolls-Royce company which was interested in measuring the internal temperature of turbine blades. Many of the detectors and experimental techniques for NRS were first developed in nuclear-physics symmetry experiments performed at Los Alamos in the late 1980s and early 1990s. The time scale for the NRS measurements is very fast: one microsecond (μs) or faster.

Examples of dynamically loaded systems studied by NRS are a metal through which a shockwave has just passed, high explosives behind the burn front after they have been detonated, or an explosively driven metal sheet jet (a jet of molten metal extruded past the neutron beam in the form of a flat sheet). The NRS temperature measurements are important because they provide temperature data that has previously been unavailable and which can lead to a better understanding of the equation-of-state for the dynamical systems under study. They also provide important tests of critical modeling calculations. In contrast to other techniques which seek to measure surface temperatures, NRS measures internal, volume temperatures.

What is a Neutron Resonance?

When a neutron that possesses an energy between 1 and 100 eV passes through a sample of atomic weight "A", at certain resonant energies the neutron can be captured by a nucleus of the sample to form an excited state in the "compound nucleus" of atomic weight "A+1". A detector placed downstream of the sample counts the neutrons that successfully pass through it. The instrument detects scintillation light from the interaction of the neutrons with lithium-6 within the detector and tags arriving neutrons by their energies; it does so by measuring the time of flight it takes for the neutrons to reach the detector.

In counting the number of neutrons versus energy, there will be absences or "resonant dips" (see Figure 1) in the observed spectra at the energies that the compound nuclei were formed. The depths of the resonances and their energies are unique to the materials the neutrons pass through. Different isotopes of the same material possess different resonances. NRS can take advantage of this uniqueness to localize temperature measurements in space by inserting a dopant that resides only in the region of interest. The shapes of these resonances depend on both intrinsic resonance properties and on the Doppler broadening produced by the motion of atoms in the target sample. By measuring the amount of broadening produced, NRS determines the temperature of the sample.

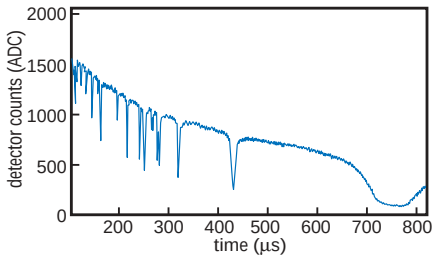

Figure 1. Transmission spectrum demonstrating resonance dips for neutrons passing through a silver sample. The dips occur at times (energies) at which neutrons are removed from the incident beam by neutron capture.

LANSCE Accelerator Provides Intense Source of Neutrons

To obtain the necessary statistics for accurate NRS temperature measurements, one requires a copious source of epithermal (1–100 eV) neutrons, and at Los Alamos, LANSCE generates the neutron source by the impinging its proton beam upon a spallation target. The linear accelerator at LANSCE accelerates protons to an energy of 800 MeV. The protons are then loaded into a circular storage ring (PSR) where they are accumulated and then released as a short, intense pulse which contains 30 trillion protons. The intense pulse of protons is directed at a uranium spallation target and, upon striking the target, produces many high-energy neutrons. These high-energy neutrons are then allowed to bounce around in a polyethylene moderator where they slow down to epithermal energies and then emerge into the NRS secondary beam line. By utilizing a specially designed uranium/polyethylene target and moderator combination, we achieve neutron flux levels an order of magnitude larger than is presently available at the main LANSCE production target.

Time Scale for Temperature Determinations

We typically place our experimental sample in the secondary beam line at a distance of 1 m from the neutron-producing target. The shorter we make this distance, the faster the temperature snapshot we can take of the dynamic state of interest. We select a specific resonance of a chosen energy to use for the NRS measurement. The time resolution of the NRS measurement is determined by the transit time of the neutrons of the chosen resonance to traverse the sample. According to the formula:

$$\Delta t \propto \frac{L\Delta E}{E^{3/2}} \quad , \qquad (1)$$

this transit time is directly proportional to the distance (L) from neutron source to sample. For the 21.1-eV resonance in tungsten-182 located 1 m from the neutron source, Figure 2 shows that the observed resonance width is 170 ns.

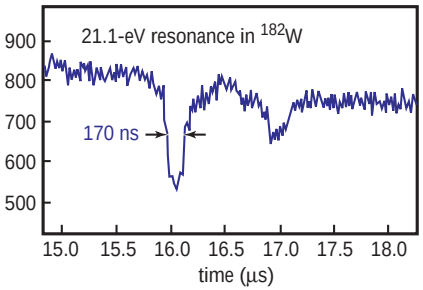

Figure 2. NRS data showing the time width of the 21.1-eV resonance in tungsten-182 for a sample located 1m away from the source of neutrons.

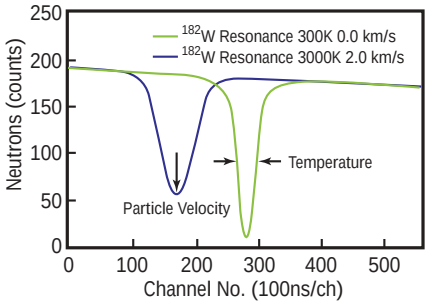

Figure 3. Expected change in the resonance line shape and position as a result of an elevation in temperature of the sample from room temperature to 3000 K and motion of the sample from being at rest to acquiring a velocity of 2km/s.

Temperature Contribution to Resonance Line Shape

For sample temperatures above the Debye temperature (see Box 1), the resonance is broadened as a result of a motion of the target atoms which can be described by Maxwell-Boltzmann theory. The resulting resonance line shape is the convolution of the intrinsic Lorentzian line shape with an energy-dependent Gaussian ($\delta_{rms} \approx \sqrt{2EkT/A}$) from the Doppler contribution. If we transform a sample to a dynamic state of elevated temperature and put it into motion as well, then the result will be both a broadening and a shift in the centroid of the probing resonance. Figure 3 illustrates this expected behavior.

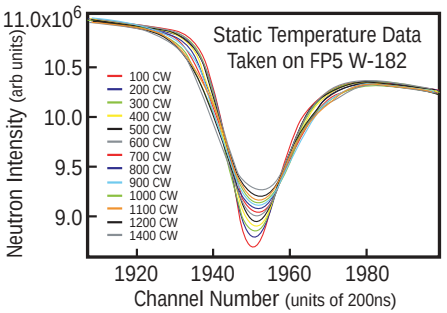

Figure 4. Measured variations in line shape for a tungsten-182 sample heated to different temperatures in a controlled oven.

Box 1

Debye temperature: The temperature q arising in the computation of the Debye specific heat, defined by kθ = hv,
where k = the Boltzman constant,
h = Planck's constant, and
v = the Debye frequency.
Also known as the characteristic temperature.

Debye specific heat: The specific heat of a solid under the assumption that the energy of the lattice arises entirely from acoustic lattice vibration modes which all have the same sound velocity, and that frequencies are cut off at a maximum such that the temperature of modes equals the numbers of degrees of freedom of the solid.

Debye frequency: The maximum allowable frequency in the computation of the Debye specific heat.

Is the temperature broadening observable? The curves shown in Figure 4 are actual measured NRS data for a tungsten sample heated to the indicated temperatures in an oven. One can easily see the progressive change in line shape as the sample temperature is increased.

In the NRS experiment, the qualitative determination of the resonance broadening is complicated by the presence of other, nontemperature-dependent factors that also affect the resonance line shape. Most prominent are the effects due to moderation of the neutrons and due to phosphorescence in the

detector. The first effect occurs because the high-energy neutrons produced in the spallation source do not always slow down to a final epithermal energy in a fixed amount of time. Rather, because of the statistical process involved, the neutrons exhibit a distribution of slow-down (moderation) times. The end result is that neutrons of the same energy emerge from the moderator over a range of time, and this spread causes a smearing of the resonance.

Phosphorescence also plays a big part in the resonance line shape. When a neutron interacts with the detector, a rapid burst of light is given off. A property of the detector's scintillation medium is to continue emitting light (phosphorescence) for a long time after the initial burst. The accumulated effect of phosphorescence is to create a false background signal under the resonances that changes their observed depth. Figure 5 compares a resonance spectrum with and without the presence of phosphorescence. The data were taken with two different detector systems, one subject to phosphorescence and one where phosphorescence was absent. Unfortunately, the one without phosphorescence cannot count rapidly enough to be used in NRS dynamic measurements. We removed the unwanted phosphorescence contribution from the data computationally.

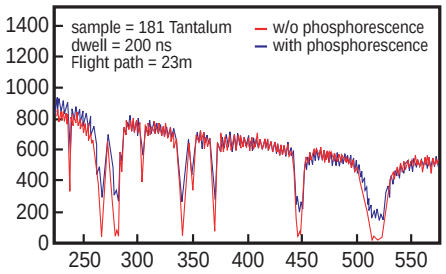

Figure 5. Comparison of resonance spectra from tantalum-181 with (blue curve) and without (red curve) the presence of phosphorescence.

A NRS Experiment: The Silver Sheet Jet

The dynamically formed silver sheet jet is one of the first systems in which NRS was used to measure temperatures. The sheet jet is formed by launching two metal liners that collide with each other to form an extruded sheet. The sheet jet passes at right angles across the neutron beam. The isotope silver-109 possesses a resonance at a neutron energy of 30.4 eV. We adjusted NRS timing so that the neutrons of this resonance passed through the sheet jet at the time of full jet formation—thus allowing broadening in the resonance to give a measure of internal jet temperature. As a reference against which to compare the dynamic experiment, we made a NRS measurement on a static silver sample of known thickness (1 mm) at a known temperature (room temperature).

Figure 6 shows the comparison between the static and dynamic resonance spectra. The plot of the sheet jet's 30.4-eV resonance in the dynamic shot is less deep because the thickness of the sheet jet is smaller than 1 mm. If one removes the influence caused by the difference in thickness between the two experiments from the line shapes, then the resonance for the silver-sheet-jet experiment is also broader than the resonance in the static experiment. A fit to the data indicates an internal sheet-jet temperature of 875 K. When we performed temperature measurements on jets formed from liners with different initial profiles, we measured varying temperatures in the range of 815–1050 K. In an attempt to better understand the observed variations in

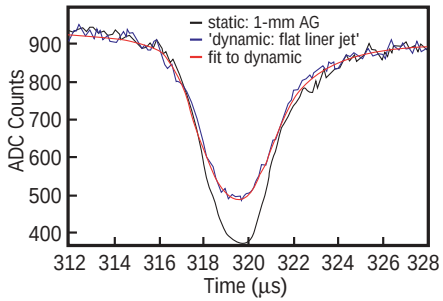

Figure 6. Comparison of 30.4-eV resonance in silver-109 for silver sheet jet (blue curve) to resonance line shape for static 1-mm sample at room temperature. Red curve shows fit to dynamic data which indicates a temperature of 875 K for a jet without variations in thickness.

temperature for the different sheet jets, we enlisted the help of the proton radiography (P-RAD) team at LANSCE . By firing the same sheet-jet shots at the P-RAD facility, we obtained proton radiographs of the sheet jet during different stages of its formation. Figure 7 shows a sequence of radiographs taken with 2 μs between radiographs. The unexpected presence of thickness variations (ripples) within the sheet jet is clearly apparent. An analysis of the data that takes the ripples into account results in a temperature that is approximately 100 K smaller than what was given by our original analysis.

Figure 7. Sequence of P-RAD radiographs that depict the evolution of the silver sheet jet. The interval between images is approximately 2 μs.

Temperature Behind the Passage of a Shock Wave in Molybdenum

An important area of research for shock-wave physics is determining the temperature behind the passage of a shockwave in a metal. Before now, no technique has been able to measure this important thermodynamic quantity for a shocked metal. We have performed NRS experiments to attempt this measurement. Figure 8 shows the set up for the NRS experiment. An aluminum flyer plate is explosively launched towards a block of molybdenum metal. The final velocity of the flyer plate (3.6 mm/μs) is great enough to initiate a shock wave in the molybdenum. At a distance between 1 mm and 2 mm into the molybdenum block, we doped a 1-mm-thick layer of the molybdenum with 1.7% tungsten-182. At 21.1 eV, the tungsten-182 possesses a resonance that can be used to measure the temperature in the doped layer immediately following the passage of the shock wave. A multipoint array of 61 detonators propels the aluminum flyer plate in a way that best maintains the flatness of the flyer plate. Approximately 0.33 μs after the flyer impacts the molybdenum, the shock wave has passed through the doped layer.

For it to be significant, the temperature measurement must be done in the short time window after the passage of the shock wave—but before the shock has had a chance to reach a boundary in the sample and to send back a release wave that alters the shocked state. NRS verifies that the measurement is made at the correct time by employing a separate laser diagnostic to observe when the shock breaks out of the back end of the molybdenum target. Figure 9 compares the 21.1-eV tungsten resonance after passage of the shock with the same resonance taken in the unshocked state. We are presently analyzing this data to extract a temperature.

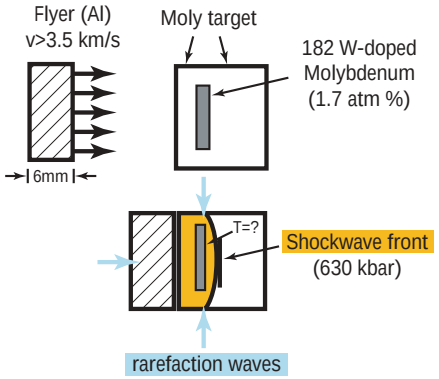

Figure 8. Cartoon showing concept for the "Temperature Behind the Passage of a Shock" experiment. An aluminum flyer impacts a molybdenum target. Inside the molybdenum target is a region doped with tungsten-182. The challenge is to measure the temperature after the shock has passed through the doped region but before rarefaction waves come in from the boundaries.

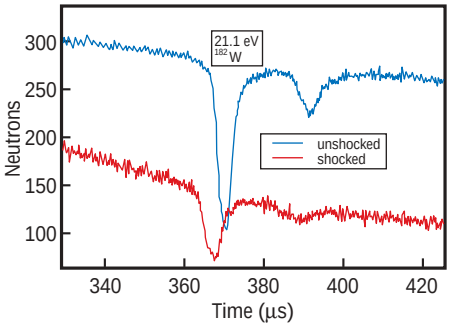

Figure 9. Resonance line shape for 21.1-eV resonance in tungsten-182 recorded before (blue curve) and immediately after (red curve) the passage of a shock wave.

Future NRS Applications

In addition to these temperature measurements, NRS has carried out scoping measurements for future experiments that will measure the temperatures at frictional interfaces in detonating high explosives. Imagine two materials which possess dissimilar sound speeds in contact with each other, as shown in Figure 10. If a shock wave is propagated (from left to right), then the different shock speeds in the two materials will cause the shock wave in one material to outrun the shock wave in the other. Further along the length of the sample there will be a region where the top material will be shocked while the bottom material will be unshocked at the same moment in time. The difference in particle velocities of the two materials will cause one to slide relative to the other; a question of interest to scientists is: what is the friction between the two surfaces? One way to get information about the friction is to measure the temperature at the interfacial surface between the two materials. We plan to isolate the interfacial surface in the region of interest by carefully placing a dopant with the resonance to be studied only in that region.

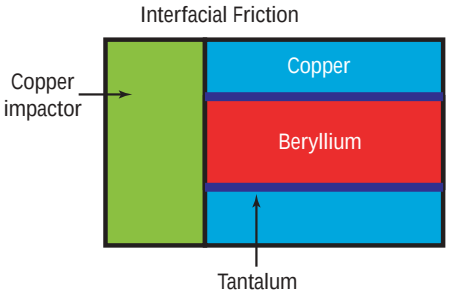

Figure 10. Cartoon showing the concept for an experiment to study interfacial friction between copper and beryllium after a copper impactor generates a shock wave in both materials. A thin tantalum layer at the interface isolates the temperature measurement to that region.

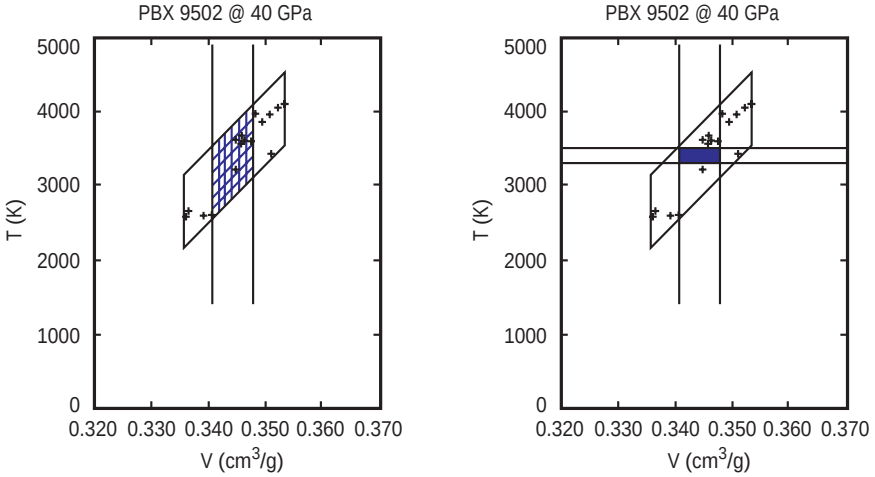

Figure 11. Improvement in knowledge of parameter space for high explosive equation-of-state that would result from a NRS temperature measurement with several hundred degrees of uncertainty.

The temperature behind the detonation wave in detonating high explosive is another case where little or no data presently exists. We plan to use NRS to measure high explosive samples doped with tungsten oxide to study the resonance broadening in the tungsten after the detonation of the high explosive has begun. Figure 11 compares the large region of uncertainty in equation-of-state space that now exists with a much more restricted region that would exist after a successful NRS measurement.

What appear to be pockets of undetonated high explosives or "dead zones" have been observed in radiographs of detonating explosives that "turn a corner." Figure 12 shows the locations of these pockets relative to the path of the detonation wave. The signature for the undetonated explosive is a difference in density from the burned regions of high explosive. But how can one confirm that these regions are truly undetonated explosive? One method is through the use of NRS. We have begun NRS scoping measurements for a set of experiments that will attempt to measure the temperature in these "dead zones" at different times in the evolution of the detonation process.

Finally, on a beamline at the Manual Lujan Center neutron production target, we have begun a series of static measurements that utilize NRS to provide important information about the individual alloy content of uranium-niobium alloys. By measuring the depth of niobium resonances in alloy samples, we are able to quantitatively determine the average niobium content in the section of alloy through which the neutrons pass. Unlike in NRS

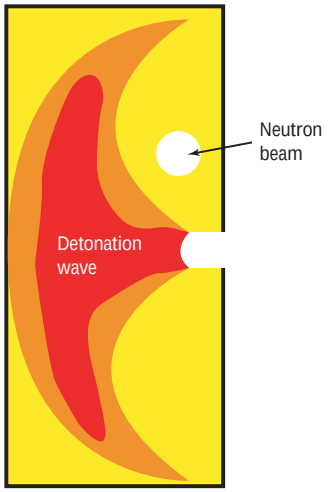

Figure 12. Schematic drawing of neutron beam used to measure temperature in a suspected "dead zone."

dynamic measurements, we perform NRS static measurements on a sample that is not changing on short time scales. Hence, the measurement uses many repetitive neutron pulses for accumulated statistics. As a reference to calibrate the measurement of a sample of unknown composition, we have made measurements on a range of alloy "sandwiches" put together from layers of prepared known amounts. The resonance curves shown in Figure 13 show the data from an alloy sample of unknown composition juxtaposed against a set of reference curves.

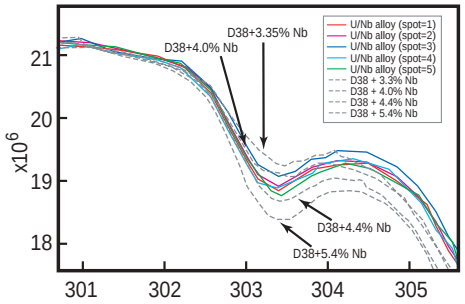

Figure 13. Change of depth of 193-eV resonance in niobium for different niobium content. Dashed lines represent reference data taken with fixed niobium content varying between 3.3 and 4.4wt%. The colored squares represent data taken with a test U-Nb alloy sample of content to be determined.

Summary

NRS offers the possibility to determine temperatures on very fast time scales in systems where, previously, temperature information has been unavailable. Numerous applications important to shock physics and materials physics have either been performed or are being planned. We have successfully measured the temperature in a sheet metal jet, and variations that we detected in the measured temperatures have led to the discovery of thickness ripples in the jet. Work that would model the temperatures is currently ongoing in another division of Los Alamos National Laboratory. NRS has obtained successfully timed data in an experiment to measure the internal temperature of a metal immediately following the passage of a shockwave. No previous experiment has yet to measure this thermodynamic quantity important to understanding the equation of state for shocked metals. Future NRS experiments are planned to understand the physics of shock-induced friction and the equation of state of high explosives after they have been detonated.

Further Reading

B. E. Crawford, J. D. Bowman, P. P. J. Delheij, *et al.*, "Neutron Resonance Spectroscopy of ^{106}Pd and ^{108}Pd from 20 to 2,000 eV," *Physical Review* C 58, 729 (1998).

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